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Improving of the filtration and regeneration performance by the SiC-DPF with the layer coating of PM oxidation catalyst

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ABSTRACT

DPF has become widely known as an indispensable after-treatment component for the purification of the particulate matter in the diesel exhaust gas. But, in order to correspond to further regulation strengthening such as carbon dioxide emission regulation and number-based particulate matter emission regulation, it must be necessary also for DPF to keep improving its performance.

In this study, It was examined how to improve both the filtration efficiency and the oxidation efficiency of PM regarding the catalyzed DPF. SiC-made 10mil/300cpsi-OctoSquare asymmetric cell structure was chosen for the DPF substrate and PM oxidation catalyst was coated on the surface of the filter wall as a layer with the device of the coating method.

As a result, it was found that the layer coated DPF has advantage on the filtration efficiency without soot accumulation and efficiency was similar to an uncoated one with 0.1 g/l soot loading. It was suggested that the layer coated DPF would satisfy the PM after-treatment with number-based viewpoint even with a clean DPF after regeneration. In addition, it was recognized that the layer coated DPF could decrease the regeneration temperature due to the improvement of the PM oxidation rate. It was confirmed that the key parameters to improve the soot oxidation by the catalyst were the filtration area of the DPF and the position of the catalyst. Both parameters improved the contact between the soot and the catalyst and the catalytic oxidation reaction proceeded faster.

INTRODUCTION

Diesel Particulate Filter (DPF) is the component to trap the Particulate Matter (PM) from diesel engine not to exhaust the PM to atmosphere, as it's name suggests. The diesel exhausted PM had been caused of the serious air pollution in past days. However, the modern diesel car is clean and no longer able to feel the PM emission from the tailpipe by equipping the DPF [1].

In European region, the emission level of both PM and NOx will be more stringent due to Euro V regulation that will be put into force in 2009. Especially, PM emission level will be one-fifth of that of Euro IV regulation. It means that DPF must become essential component for almost all diesel cars. In addition, not only weight-based but also number-based PM emission regulation is also under discussion to introduce because finer particle is thought to be more harmful on the human body [2]. Moreover, the introduction of CO₂ emission regulation is now considered seriously as a countermeasure against global warming by the greenhouse effect. Of course DPF must be required to improve its performance continuously to make the diesel car "clean" and "green" more and more.

Equipping the DPF adversely affects the fuel economy of diesel car more or less. In other words, it makes CO₂ emission higher compared with DPF unequipped car. One main reason is the backpressure increasing by the accumulation of PM on the DPF. And the other is the additional energy consumption to burn the accumulated PM and regenerate the DPF periodically [3]. It is considered that to improve the PM oxidation performance of the DPF will be effective as a measure to minimize these energy losses. At first, this is because the pressure loss will increase gently if the soot can be oxidized simultaneously while the soot is accumulating on the DPF. Second, this is because it will be needed less energy for the DPF regeneration operation if the DPF can be regenerated by lower inlet gas temperature and shorter time.

The PM oxidation performance can be increased by the catalyst. A lot of research works are proceeding with catalytic composition, mainly oxide base, to improve the catalytic oxidation of the PM. It is also known by some research works that the improvement the contact probability between the soot and the catalyst is one of the key factors for the catalytic PM oxidation because PM is the solid form, unlike CO, HC and NOx which is the gas form [4][5]. However, there are few examples which are examined about the coating position of the PM oxidation catalyst on the DPF.

Now, by observing the condition of the PM accumulation of the commonly-used DPF, it is recognized that the PM is trapped at the surface of the wall (see Fig.1). Therefore, the catalyst only which is located at the surface of the wall can contact with the PM flowed into the DPF. Two measures can be thought to improve the contact between the PM and the PM oxidation catalyst on the catalyzed DPF. One is to extend the filtration area of the DPF as much as possible by modifying the cell structure. The other is to increase the amount of the catalyst particle which is located at the surface of the wall.

Regarding number-based PM emission regulation, to improve the filtration efficiency especially about the fine particles is required. As mentioned above, the PM is mostly trapped at the surface of the wall of the DPF and is deposited on the wall with forming the layer. The filtration efficiency becomes close to 100% after certain thickness of this soot layer is formed. This is because the formed soot layer plays a role in trapping the soot which is exhausted from the engine later.

In this study, the examination of the new catalyzed DPF which was developed to improve both the PM oxidation performance and the filtration efficiency was performed based on the consideration described above. Evaluated catalyzed DPF was coated the PM oxidation catalyst which has similar size with PM only on the surface of the wall with the layer form. It is reported that the evaluation results of the SiC-DPF with the layer coating of PM oxidation catalyst regarding the soot oxidation performance and filtration performance.

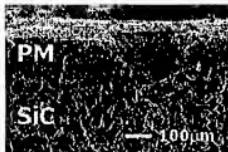


Fig.1 Soot collection state on SiC-DPF

EXPERIMENTAL

Fig.2 shows the relationship between the filtration area and the relative density of the filter. There are two ways to increase the filtration area, one is by increasing cell density from 200 cpsl to 300 cpsl, and the other is by changing cell design from square shape to octagonal shape about the inlet cell. Latter cell design, so called "OS cell", has higher ash storage capacity due to the larger inlet cell volume per filter volume [6]. In addition, it has larger effective filtration area per filter volume, because slanted wall between two inlet cells can also be effective for filtration. Two kinds of cell structure of re-crystallized SiC-DPF were tested in this study. Both cell structures has 10mil (0.25mm) of wall thickness and about 300cpsl of cell density and one has commonly-used square cell design and the other has OS

cell design [7]. Regarding pore structure, both have 11 micron of pore diameter and 42% of porosity.

Catalyst particle was coated on the surface of the wall of above DPF substrates. In this study, CeO₂ based material with about 300nm of particle size was chosen as the catalyst. No precious metal was used. The coating amount of catalyst was about 15g/L. Fig.3 shows the cross cut SEM image about test sample. The catalyst was coated on the surface of the wall uniformly including the slanted wall with about 10 micro of thickness.

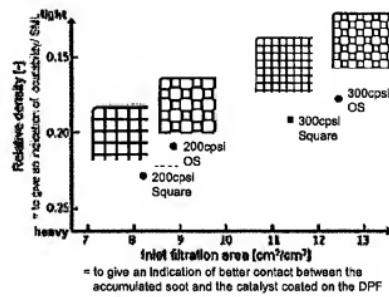


Fig.2 Properties about several cell designs

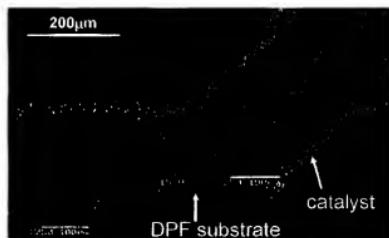


Fig.3 Layer coating on the DPF wall

Table 1 shows the factors and levels of this study. SiC-DPF with the layer coating of PM oxidation catalyst was evaluated about following two functionalities, compared with uncoated DPF.

1. Pressure Loss

Two kind of measurement were performed about the pressure loss after catalyst coating. One was the cold flow pressure loss. Room temperature air was made to flow through a clean filter with an apparent cross-sectional area of 1180 mm² (34.3 mm-square), and then, the difference between the pressure at the filter inlet and that at the outlet was measured. The other was the transient pressure loss behavior during soot loading. The engine exhaust gas was distributed and made to flow through a filter for certain hours. The distributed gas flow rate was controlled so that the cross-sectional flow rate was about 5 m/s.

2. Soot Oxidation Rate

Table 2 shows the factors and levels of the soot oxidation rate measurement test. This test was conducted to evaluate the effect of the layer coating of the PM oxidation catalyst on the oxidation rate of the PM collected in the DPF according several temperatures. A soot generator was used in collecting the soot. The soot from the soot generator was collected in the DPF so as to be 2 g/l. A homemade apparatus was used in this test. The soot in the DPF was oxidized by a gas heated with an electric heater (see Fig.4). The gas consisted of air diluted with nitrogen so that O₂ concentration would be 10%. No NOx was included in the gas. Gas space velocity was fixed at 72,000 h⁻¹ at room temperature. Regeneration temperature was defined as the temperature at 15 mm from the filter inlet. First, the DPF in which the soot was loaded was heated up to 700 °C under constant N₂ flow and maintained for 10 minutes in order to minimize the influences of the SOF composition in each test and study the soot oxidation plainly. Afterwards, it was lowered to a predetermined regeneration temperature, and the soot oxidized by switching to 10% O₂. A history of the temperature in the filter was measured by thermocouples and the amount of CO and CO₂ generated were recorded using a SHIMADZU CGT-7000 system. The rate of oxidation was derived from measured CO and CO₂ values. In addition, Arrhenius plots were made by plotting soot oxidation rates against temperature, and the apparent activation energy was calculated from the gradients of these lines.

3. Filtration Efficiency

Number-based filtration efficiency was measured about the test samples in Table 1. PSA DW10 (2.0L, turbo charged common rail engine) was run at 3000 rpm/50 Nm and exhaust gas was by-passed to the measuring device. The by-passed gas was diluted by the device to 1/1000 by a simulated gas. A diameter and number of the PM particles was measured in front of and behind the SiC-

DPF by SMPS and filtration efficiency was calculated by the difference of the number of the PM particles between the DPF for each particle diameter.

Table 1. Factors and Levels of the test sample

Factor	Level
DPF substrate	10mil/300cpsl-11μm/42%-Square
	10mil/300cpsl-11μm/42%-OS
Catalyst coating	Uncoated
	Catalyzed
Sample size	34.3mm-sq. x 150mm

Table 2. Factors and Levels of the soot oxidation test

Factor	Level
Regeneration temperature	450°C, 500°C, 530°C, 560°C, 590°C, 620°C
Initial soot load	2 g/l
Gas composition	10%-O ₂ /90%-N ₂ *No NO and NO ₂

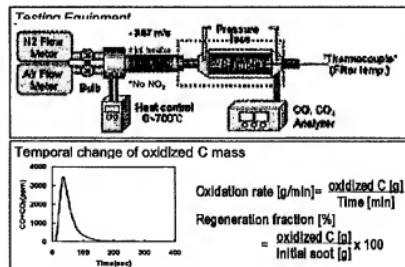


Fig.4 Soot oxidation rate measurement test

RESULTS

PRESSURE LOSS AFTER LAYER COATING

Fig.5 shows the test result on pressure loss vs flow rate. Pressure loss wasn't increased a lot by the layer coating. In general, the pressure loss was increased by the catalyst coating when the certain amount of pores of the DPF was closed by the coating [8]. The pressure loss was less increased in the case of OS cell compared with Square cell. It was supposed that OS cell had thinner catalyst layer when same amount of catalyst was coated because of larger filtration area and the gas was easier to go through.

Fig.6 shows the test result on the transient pressure loss during soot loading. The pressure loss was increased rapidly in the case of uncoated filter when the soot was penetrated into the pore and thick soot layer was built inside and on the surface of the filter wall. As observed in Fig.3, the layer coating was formed on the surface of the wall and the pore of the DPF wasn't closed, so the coated filter has less soot penetration than uncoated one and therefore the pressure loss increased relatively linearly.

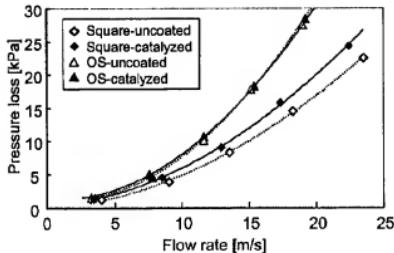


Fig.5 Cold Flow Pressure Loss

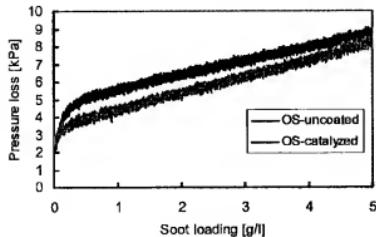


Fig.6 Soot Loading vs Pressure Loss

SOOT OXIDATION RATE

Fig.7 and Fig.8 shows the transient CO and CO₂ generation during soot oxidation at 560 °C about OS-uncoated sample and catalyzed one. The catalyzed samples test result in Fig.8 has one order of magnitude greater in Y-axis, so it is recognized that the catalyzed sample has about 10 times higher soot oxidation rate at same regeneration temperature.

According CO₂ selectivity, the average CO₂ selectivity was about 50% in the case of uncoated sample. It means 50% of the soot converted to CO₂, not CO. On the other hand, the catalyzed sample had above 80% of CO₂ selectivity at same regeneration temperature. It was found that more than 80% of CO₂ selectivity could be achieved even if there is no PGM in the catalyst.

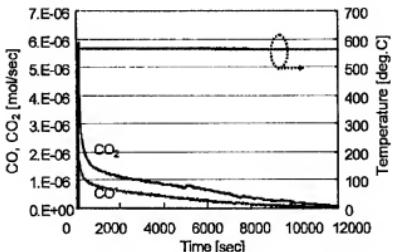


Fig.7 CO/CO₂ during regeneration @560°C
(OS, uncoated)

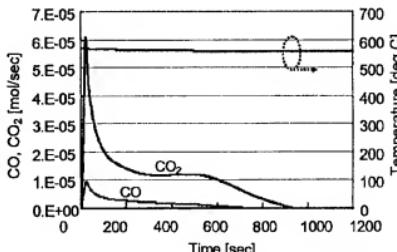


Fig.8 CO/CO₂ during regeneration @560°C
(OS, catalyzed)

Fig.9 shows the transient regeneration efficiency during soot oxidation at 560 °C. OS cell has higher regeneration speed compared with Square cell, especially after the layer coating. OS cell design has about 10% larger filtration area compared with Square cell design. So the contact between the soot and the catalyst on the surface of the wall is improved about 10% basically. Though when it is focused on the regeneration efficiency at 500sec, OS cell was completely regenerated and Square cell was only 50% regeneration of initial soot amount. It is supposed that the upper soot of the soot layer dropped onto the catalyst layer when the soot at the border with the catalyst burn and chain catalytic oxidation reaction occurred. It was suggested that increasing of the filtration area by devising the cell structure like as OS cell could be effective for improving the soot oxidation rate when the layer coating was applied.

Fig.10 shows Arrhenius plots of the soot oxidation rate with several regeneration temperatures. Within the temperature range from 500 °C to 650 °C, the layer coating gives more than 10times faster soot oxidation rate. It was suggested that the regeneration could be done with lower temperature and shorter time by applying the layer coating and produced smaller amount of CO₂ during forced regeneration. Compared between

Square cell and OS cell, there are no difference above 600 °C (1000/T=1.14) and big difference below 600 °C. In general, thermal oxidation is dominant above 600 °C and catalytic oxidation is dominant below 600 °C to the soot oxidation. It was confirmed the layer coated catalyst could be much active when the filtration area was larger because of the contact between the soot and the catalyst.

FILTRATION EFFICIENCY

Fig.11 and Fig.12 shows the number-based filtration efficiency measured by SMPS. Regarding OS uncoated filter, the average filtration efficiency was 80% and about 100nm of particle diameter wasn't collected by the filter with high efficiency when the soot wasn't loaded on the filter. When 0.1 g/l of the soot accumulated, the filtration efficiency increased to the high level and it didn't have the particle diameter dependence. The layer coated catalyzed DPF had more than 95% of the filtration efficiency even if the soot wasn't accumulated. It was found that the filtration performance of the layer coated DPF would be equal with the 0.1 g/l of soot accumulated uncoated one, because the similar morphology of the surface of the filter wall was formed not by the soot but catalyst particles.

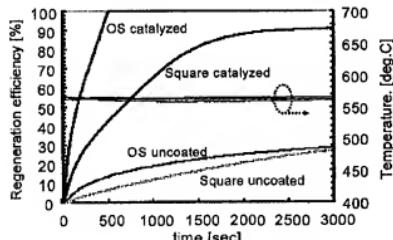


Fig.9 Regeneration efficiency @ 560°C

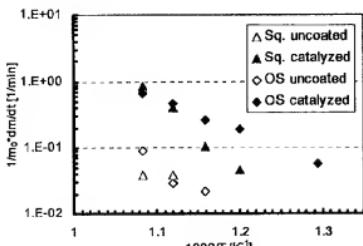


Fig.10 Arrhenius Plots

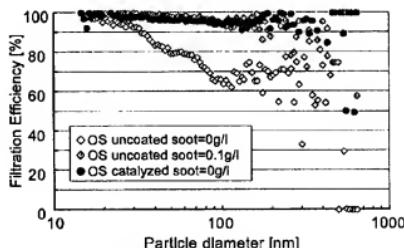


Fig.11 Filtration Efficiency by SMPS

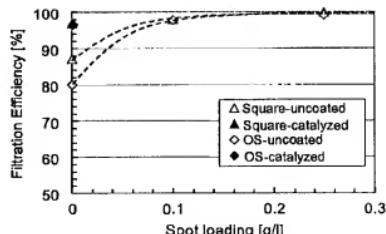


Fig.12 Soot Loading vs Filtration Efficiency

CONCLUSIONS

To satisfy the future regulation about PM and CO₂ reduction, the SiC-DPF with layer coating of PM oxidation catalyst was developed and evaluated on fundamental performances of DPF, those are the pressure loss, the filtration efficiency and regeneration. As a result, the following knowledge was obtained:

1. The increase of the pressure loss due to the catalyst coating with the layer was within 10% of the uncoated DPF. It had relatively low impact on the pressure loss because the DPF wall pores were not closed by the coating and the porous layer by the catalyst particles was thin.
2. The layer coated catalyzed DPF showed high soot oxidation performance especially about OS cell design one. It was confirmed that the key parameters to improve the soot oxidation by the catalyst were the filtration area of the DPF and the position of the catalyst. Both parameters improved the contact between the soot and the catalyst and the catalytic oxidation reaction proceeded faster.
3. Although SiC-DPF itself has high filtration efficiency the filtration efficiency improved with the catalyst layer. It had near 100% filtration without soot accumulation and efficiency was similar to an uncoated OS DPF with 0.1 g/l soot loading. It was suggested that the layer coated DPF would satisfy the PM after-treatment with number-based viewpoint even with a clean DPF after regeneration.
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